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Phys. Rev. Lett. **106**, 183902 — Published 3 May 2011

DOI: [10.1103/PhysRevLett.106.183902](https://doi.org/10.1103/PhysRevLett.106.183902)

## Saturation of the all-optical Kerr effect

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Saturation of the intensity dependence of the refractive index is directly computed from ionization rates via a Kramers-Kronig transform. The linear intensity dependence and its dispersion are found in excellent agreement with complete quantum mechanical orbital computations. Higher-order terms concur with solutions of the time-dependent Schrödinger equation. Expanding the formalism to all orders up to the ionization potential of the atom, we derive a model for saturation of the Kerr effect. This model widely confirms recently published and controversially discussed experimental data and corroborates the importance of higher-order Kerr terms for filamentation.

PACS numbers: 42.65.Tg, 42.65.-k, 52.38.Hb, 42.68.Ay

Most nonlinear optical effects can be understood in the perturbative limit with two or three interacting optical waves, giving rise to contributions  $\chi_{ij}^{(2)} E_i E_j$  or  $\chi_{ijk}^{(3)} E_i E_j E_k$  to the polarization  $P$ , respectively. Higher-order  $\chi^{(n)}$  ( $n \geq 4$ ) terms can be formally considered. Yet, a perturbative description of higher order effects is rarely useful as often enough a large number of waves interact simultaneously as, for example, in high-harmonic generation [1]. The role of  $\chi^{(5)}$  effects for arresting catastrophic optical self-focusing has been discussed already more than 20 years ago [2–4].  $\chi^{(3)}$  effects, namely the all-optical Kerr effect, give rise to an increase of the refractive index with intensity  $n = n_0 + n_2 I$  and a resulting focusing nonlinear lens. A  $\chi^{(5)}$  dependence with negative sign and defocusing nonlinear lensing would explain the phenomenon of filamentation, i.e., the formation of long self-guided light strings with nearly constant diameter. Early experimental results indicated that filamentation cannot only be explained by plasma formation [5], which gives rise to a negative index contribution suitably described by Drude theory. Nevertheless, refined theoretical models succeeded in explaining even complex experimental results without the need for including a saturation of the Kerr effect [6, 7]. Recently, this accepted picture was challenged by measurements [8, 9] that indicate yet again a strong influence of higher-order nonlinearities to the extent that filament formation is explained in the complete absence of plasma formation. These results have been controversially discussed [10, 11]. In the following, we provide an independent and previously unreported approach towards computing Kerr saturation. Our approach is based on a Kramers-Kronig transform [12] of optical absorption derived from Keldysh theory [13]. This analysis supports the experimental results in Ref. [8], indicating that we may have in fact a paradigm shift in explaining femtosecond filamentation [10].

Our model is based on a recent modification [13] of Perelomov-Popov-Terent'ev (PPT) theory [14], the former providing cross-sections for multiphoton ionization

according to

$$\sigma_K(\omega) = \frac{2\sqrt{2}C^2}{\pi} (2e)^{2n^*} \left(\frac{e}{2}\right)^{2K} \omega_p^{-3K+1} \left(\frac{q_e^2}{\hbar m_e \epsilon_0 c}\right)^K \times \left(\frac{\omega_p}{\omega}\right)^{2n^*+2K-\frac{3}{2}} \exp\left(-\frac{\omega_p}{\omega}\right) w_0 \left[\sqrt{2K - 2\frac{\omega_p}{\omega}}\right] \quad (1)$$

for  $\omega \geq \omega_p/K$ . Here  $\omega$  is the optical frequency,  $q_e$  and  $m_e$  denote electron charge and mass, respectively,  $\epsilon_0$  is the vacuum dielectric constant, and  $c$  denotes vacuum light speed.  $\hbar\omega_p$  is the ionization potential of the gas species under consideration. The constant  $K = \langle \omega_p/\omega + 1 \rangle$  counts the number of photons required for ionization, where  $\langle x \rangle$  denotes the integer part of  $x$ . The effective principal quantum number of the bound state is given by  $n^* = Z\sqrt{\omega_H/\omega_p}$ , where  $\hbar\omega_H = 13.6\text{eV}$  is the Rydberg energy and  $Z$  is the charge number of the ionic residuum. The constant  $C = 2^{n^*-1}/\Gamma(n^* + 1)$  is related to the asymptotic expansion of the ground-state electronic wavefunction, and  $w_0[x]$  denotes Dawson's function [15]. From the cross-sections  $\sigma_K$ , the  $K$ -photon absorption coefficients  $\Delta\alpha_K$  may then be calculated according to  $\Delta\alpha_K(\omega) = K\hbar\omega\rho_{\text{nt}}\sigma_K I^{K-1}$ , with a particle density  $\rho_{\text{nt}} = 2.7 \times 10^{19}\text{cm}^{-3}$  corresponding to standard conditions. For atomic argon, this perturbative approximation is expected to hold for intensities up to  $50\text{TW/cm}^2$ . This intensity corresponds to a Keldysh-parameter of  $\gamma = 1.62$ , where MPI is the dominant ionization channel [16].

Kramers-Kronig theories have been successfully applied to nonlinear refraction in solids [12, 17]. Here we combine this method with Keldysh theory to compute nonlinear refraction in inert gases. In principle, as pointed out in [12], the use of Kramers-Kronig relations requires knowledge of nondegenerate multiphoton absorption coefficients  $\Delta\alpha_K^N(\omega_1, \dots, \omega_K)$ . These coefficients cannot easily be provided by PPT theory. Instead,

we use

$$\Delta\alpha_K^N(\omega_1, \omega_2, \dots, \omega_K) = \Delta\alpha_K \left( \frac{\omega_1 + \dots + \omega_K}{K} \right), \quad (2)$$

as an estimate, generalizing the model successfully used to compute  $n_2$  in solids [17].

Using this proven simplification, we find that the nonlinear refraction coefficients  $n_{2k}$  are related to  $(k+1)$ -photon absorption coefficients  $\sigma_{k+1}$  via the KK relations according to

$$n_{2k}(\omega) = \frac{\hbar c \rho_{\text{nt}}}{\pi} \mathcal{P} \int_0^{\infty} (\Omega + k\omega) \frac{\sigma_{k+1} \left( \frac{\Omega + k\omega}{k+1} \right)}{\Omega^2 - \omega^2} d\Omega, \quad (3)$$

where  $\mathcal{P}$  denotes Cauchy's principal value integral. It has been shown in Ref. [17] that Eq. (2) is a reasonable approximation when AC Stark terms are neglected in the model. As a benchmark for our model, we calculate the leading term  $n_2(\omega)$  of nonlinear refraction for helium [Fig. 1], setting  $k = 1$ .

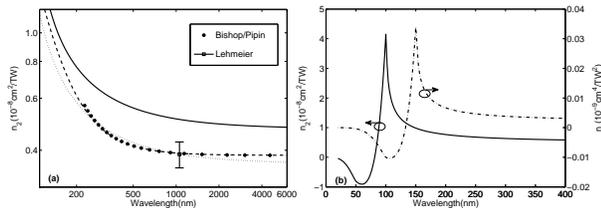


FIG. 1: (a) Nonlinear refractive index  $n_2$  of He below the 2PA resonance at  $\lambda = 85$  nm. Solid lines:  $n_2$  dispersion as extracted from Eq. (3). Dashed line: fit of theoretical data to scaling law (dots) from [21]. (b) Same in the vicinity of the resonance (solid line), dashed-dotted line showing  $n_4(\omega)$ .

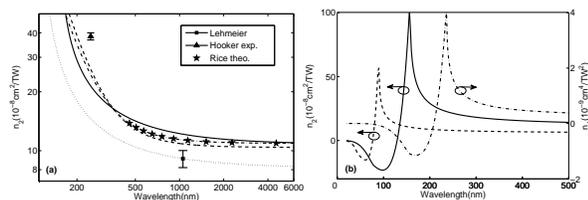


FIG. 2: (a) Nonlinear refractive index  $n_2$  of Ar from Eq. (3) below the 2PA resonance. Dash-dotted line: power series in  $\omega^2$  [21, 22] fitted to theoretical data [22] (stars). Dashed line: experimental  $n_2$  data [21] Dotted line: Lehmeier data extrapolated with scaling law given in [19]. (b)  $n_2$  of neutral argon in the vicinity of the 2PA resonance at  $\lambda = 85$  nm [solid line, Eq. (3)]. Dashed line:  $n_2$  of singly ionized  $\text{Ar}^+$ . Dashed-dotted line:  $n_4(\omega)$  for Ar.

Among the inert gases, helium is the least complex atom and the only one for which detailed computations of  $n_2$  from atomic wave functions exist. For large

wavelengths, our analysis indicates a value  $n_2 = 4.8 \times 10^{-9} \text{ cm}^2/\text{TW}$ , which deviates by only 26% from the value  $3.8 \times 10^{-9} \text{ cm}^2/\text{TW}$  that was derived in [18] using explicitly electron-correlated wave functions. Keeping in mind that the absorption spectra  $\sigma_K$  have been derived from strong field ionization rates for which often an order of magnitude agreement with experimental data is considered reasonable, our Kramers-Kronig approach provides an excellent prediction of  $n_2$ . Our model also correctly reproduces the dispersive behavior of  $n_2$  predicted in Ref. [18] and reasonably agrees with experimental data at 1.06  $\mu\text{m}$  wavelength [19]. Even better agreement is obtained for argon [Fig. 2]. Going beyond the tabulated values of [18] for helium, our computations predict that  $n_2$  reaches a maximum value  $n_2 \approx 4 \times 10^{-8} \text{ cm}^2/\text{TW}$  at about 100 nm wavelength, which corresponds to half the ionization energy. Going to even smaller wavelengths,  $n_2$  crosses zero at 85 nm and stays negative up to the ionization energy equivalent of 50.4 nm wavelength. This prototypical behavior with a sign change at approximately 60% of the ionization threshold is seen for all inert gases and duplicates the dispersion characteristics of  $n_2$  in solids [17]. Compared to our previous work [20], the possibility to extend  $n_2$  computation beyond half the ionization energy into the negative region and the correct prediction of its dispersion in the positive region both arise from usage of the improved ionization cross sections provided in Ref. [13].

Similarly good agreement of Eq. (3) with independent experimental and theoretical data is obtained for neon, krypton and xenon, for which our approach reproduces experimental and theoretical data [21, 22] within 15% precision.

Figure 2 shows an  $n_2$  computation for argon showing similar features as the helium example. Given that the ionization energy of argon is only 15.76 eV and that a smaller number of photons is required to reach the continuum,  $n_2$  of argon is about a factor of 20 larger in the infrared, with a value of  $\approx 10^{-7} \text{ cm}^2/\text{TW}$ . This value agrees favorably with commonly used reference data [19] and was also reproduced by Loriot *et al.* in their measurements. Compared to helium, the zero crossing of  $n_2$  is now shifted to a wavelength of 140 nm.

$n_2(10^{-8} \text{ cm}^2/\text{TW})$	$Z = 1$	$Z = 2$	$n_2(\omega \rightarrow 0)$	Refs.[18, 21]
He	0.52	0.03	0.48	0.38
Ne	1.31	0.27	1.18	0.96
Ar	12.68	6.14	10.84	10.40
Kr	30.69	17.28	25.63	23.17
Xe	91.58	55.17	73.87	61.39

TABLE I: Nonlinear refractive index  $n_2$  at 800 nm for Ar and  $\text{Ar}^+$  ( $Z = 1$ ), ( $Z = 2$ ) and Ar in the static limit  $n_2(\omega \rightarrow 0)$ . Experimental data (rightmost column) as compiled from [18, 21] and corrected for the dispersion of the DFWM process with Eq. (13) in [20].

Nonlinear refraction, in principle, holds two potential mechanisms for saturation. First, the generation of free electrons will replace a number of neutral atoms by ions. In the case of  $\text{Ar}^+$ , this raises the ionization threshold to 27.63 eV, which, in turn, has to reduce the resulting  $n_2$  values as illustrated by the transition from argon to helium. A computation of the resulting values for  $\text{Ar}^+$  is shown as the dashed line in Fig. 2(b), which confirms a reduction of  $n_2$  by a factor of  $\approx 2$ . Table I lists computed values of  $n_2$  of all noble gases and their first ionic species ( $Z = 1, 2$ ) for 800 nm wavelength.

While this first mechanism suggests a depletion of the neutral atoms as the cause for saturation, the same effect may also occur due to higher-order Kerr terms  $n_{2k}$  with  $k \geq 2$ . These terms can also be computed using our theoretical approach. As an example, we have plotted  $n_4(\omega)$  of helium and argon as dashed-dotted lines in Figs. 1(b) and 2(b), respectively. According to our model, argon displays an infrared limit  $n_4(\omega \rightarrow 0) = 2 \times 10^{-12} \text{ cm}^4/\text{TW}^2$ , reaches a 12 times higher maximum value at 240 nm, and is negative below 210 nm. This behavior is again in agreement with the prototypical dispersion of the Kerr coefficients, yet caused by three-photon rather than two-photon absorption.

There is an apparent contradiction to the negative value  $n_4 = -0.36 \pm 1.03 \times 10^{-9} \text{ cm}^4/\text{TW}^2$  reported in Ref. [8]. In principle, a negative  $n_4$  value at 800 nm appears to be incompatible with the dispersion of the Kerr terms predicted by our model.

The analysis of higher-order Kerr terms can easily be continued to arbitrary order  $k$  in our model, even beyond the highest-order experimental  $n_{10}$  term in Ref. [8]. Again, helium provides a benchmark for this extension. Using the theoretical susceptibility data published in Ref. [23], we find excellent (within 10%) agreement with  $n_4$  and  $n_6$  values derived from Kramers-Kronig theory. The agreement only breaks down in the highest-order coefficient  $n_{10}$  extracted from the series expansion used in Ref. [23].

For argon at a wavelength of 800 nm, we compute positive coefficients up to  $n_{18}$ . Higher-order coefficients are negative and merge into a series with constant ratio between successive coefficients [24]. This geometric series ensures convergence up to 70  $\text{TW}/\text{cm}^2$ . Despite the nonalternating structure of our sequence of Kerr coefficients, we find a good agreement between the computed nonlinearly induced index change  $\Delta n_{\text{Kerr}} = n_2 I + n_4 I^2 + n_6 I^3 + \dots$  and the experimental values for argon in [8], see Fig. 3(a). Our model predicts an increase of  $\Delta n_{\text{Kerr}}$  up to about 42  $\text{TW}/\text{cm}^2$  and inversion of the index change at 49  $\text{TW}/\text{cm}^2$ . This is contrasted by experimental values of 30  $\text{TW}/\text{cm}^2$  and 34  $\text{TW}/\text{cm}^2$ , respectively. Apart from this apparent scaling issue, both curves agree remarkably well within the error bounds. These bounds have been extracted from the uncertainty of the experimental Kerr-coefficients given in Ref. [8]. In

contrast, the error of our theoretical values has been estimated from the comparison of our benchmark  $n_2$  values with independent values in the literature.

From the coefficients for He, Ne, Kr and Xe shown in [24], we deduce inversion intensities of 112, 89, 40, and 30  $\text{TW}/\text{cm}^2$ , respectively. Our analysis also qualitatively agrees with solutions of the time-dependent Schrödinger Equation for atomic hydrogen [25]. Agreement with corresponding calculations for argon performed in Ref. [26] are shown by the dashed-dotted curve in Fig. (3), indicating a favorable agreement of the inversion intensity within reasonable error margins. For the air constituents  $\text{O}_2$  and  $\text{N}_2$ , Figs. 3(b) and (c) show the experimentally measured  $\Delta n_{\text{Kerr}}(I)$  in comparison to our theoretical derivations. In order to apply the PPT model to molecular gases, we employ the semi-empirical model of Ref. [27] with  $Z_{\text{eff}} = 0.53$  and 0.9 for  $\text{O}_2$  and  $\text{N}_2$ , respectively. For oxygen, our theory yields a long-wavelength limit of  $n_2(0) = 0.7 \times 10^{-7} \text{ cm}^2/\text{TW}$ , which agrees excellently with the electronic contribution of  $n_2(0) = 0.746 \times 10^{-7} \text{ cm}^2/\text{TW}$  computed in [28].

Despite of delivering higher inversion intensity than reported in [8], our model nevertheless confirms plasma clamping to occur at significantly higher intensities than Kerr saturation. Figure 3(a) shows the refractive index change  $\Delta n(I) = n_2 I - \rho/2\rho_c$  induced by the generation of free electrons with density  $\rho$  under experimental conditions of [8].  $\rho_c = m_e \epsilon_0 \omega^2 / q_e^2$  is the critical plasma density. Clearly, plasma clamping is expected at intensities beyond  $\approx 100 \text{ TW}/\text{cm}^2$ , i.e., well above Kerr saturation.

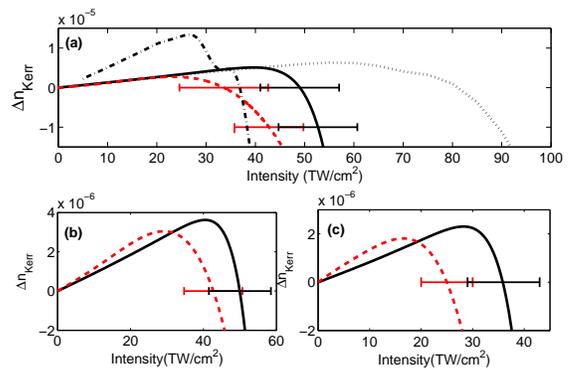


FIG. 3: Kerr saturation in (a) argon, (b) nitrogen and (c) oxygen at 800 nm due to higher order Kerr terms [Eq. (3), solid lines], classical filamentation model due to plasma clamping (dotted line), and experimental results [8] (dashed lines). Dashed dotted line in (a) depicts TDSE results for argon found in [26].

In a second simulation, we model the potential effect of depletion of neutral atoms on the index change. Given a fraction  $p$  of ionized atoms, we compute  $n_2(I) = p n_{2,\text{Ar}^+} + (1 - p) n_{2,\text{Ar}}$  where  $p$  is computed under the assumption of 90 fs Gaussian pulses using the ioniza-

tion model of [13], duplicating experimental conditions of Ref. [8]. In this case we yield a classical saturation behavior (not shown), yet at much higher intensities  $I > 250 \text{ TW/cm}^2$  and without any index inversion. Complete ionization of argon requires intensities of  $\sim 300 \text{ TW/cm}^2$ , and even then only a 50% index change from  $n_2 = 1.3 \times 10^{-7} \text{ cm}^2/\text{W}$  at  $p = 0$  to  $n_2 = 0.6 \times 10^{-7} \text{ cm}^2$  at  $p = 1$  results. As this happens nearly an order of magnitude beyond the inversion intensities discussed previously, depletion effects can be clearly ruled out.

These results shed new light on the long-disputed mechanism behind filament formation. First and foremost, saturation of the Kerr effect cannot be explained by inclusion of the next higher-order coefficient  $n_4$  alone. Instead, similar as in the transition from third-harmonic generation to high-harmonic generation, many coefficients start to act simultaneously. Nevertheless, the assumptions of perturbative nonlinear optics are expected to hold as long as the intensity does not exceed the validity of the MPI regime. Beyond that regime, a perturbative expansion of the ionization rate provided by PPT ceases to exist.

While a true depletion-caused saturation  $n_2(I) = n_2(0)/(1 + I/I_{\text{sat}})$  can be developed into a Taylor series in  $I$  resulting in a sequence of  $n_{2k}$  with alternating signs, our model predicts that all  $n_{2k}$  are positive until the driving  $(k + 1)$ -photon process reaches about 75% the ionization energy. This causes a nearly unperturbed linear increase of the index change  $\Delta n_{\text{Kerr}}$  up to a certain threshold. Above this threshold,  $\Delta n_{\text{Kerr}}$  will rapidly decrease and reach strong negative values. Comparing the absolute values of  $n_4$  from our model with Ref. [8], we generally compute smaller values than found in Taylor series analysis of experimental data. This finding corroborates that saturation of the Kerr effect may be perfectly compatible with experimentally observed efficiencies of fifth-order harmonic generation processes [10].

Despite its slightly different functional shape, our results qualitatively confirm the saturation behavior suggested by Loriot *et al.* This agreement strongly suggests to include a saturation mechanism into future models of filament formation. Modeling of white-light propagation, however, may turn out to be difficult because of the strong dispersion of the higher-order coefficients, and methods for efficient modeling of dispersive nonlinearities may have to be found. We believe that this work has important consequences for nonlinear optics in a large class of materials, including gases, solids, and metamaterials. In fact, this may truly induce a paradigm shift in the understanding of filamentation.

Financial support by the Deutsche Forschungsgemeinschaft, grants DE 1209/1-2 and STE 762/7-2, is gratefully acknowledged. GS gratefully acknowledges support

by the Academy of Finland (project grant 128844).

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